

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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IN THE APPLICATION OF

LAIMUTE R. SVARCAS, WILLIAM K.S. CLEVELAND, AND JOHN L. PETRIC

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EXAMINER: V. VASISTH

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GROUP ART UNIT: 1797

TITLE: LUBRICANT COMPOSITION SUITABLE FOR DIRECT FUEL INJECTED,  
CRANKCASE-SCAVENGED TWO-STROKE ENGINES

Wickliffe, Ohio

Hon. Commissioner for Patents  
P. O. Box 1450  
Alexandria, VA 22313-1450

Sir:

**DECLARATION UNDER 37 C.F.R. §1.132**

I, Ewan Delbridge, declare as follows:

I received a Bachelors of Science (with Honours) degree in the field of Chemistry (with a minor in Pharmacology) in 1995 from Monash University, Australia, and a Doctor of Philosophy degree in the field of Synthetic Chemistry in 1999 from Monash University, Australia.

I have been employed by The Ohio State University (2000-2003) and University of North Dakota (2003-2007) as a Postdoctoral Researcher and Assistant Professor of Chemistry, respectively, and am currently employed by The Lubrizol Corporation (2007-present date). I am the author or co-author of several refereed publications in international journals. At The Lubrizol Corporation, I am currently responsible for: a) designing, synthesizing and implementing new lubricant componentry which imparts enhanced performance to lubricant formulations; b) understanding the correlation between molecular architectures and fluid performance; c) understanding componentry synergies and antagonisms in formulations.

I am familiar with the invention claimed in the above-mentioned case. Upon request from Dr. Patrick Mosier and in order to illustrate one of the features of one of

the materials prepared in the Blythe reference, U.S. 5,264,005, cited in the prosecution of the above-identified application, the following experiments were performed closely under my direct supervision:

*Identical* reproduction of Example A-8 of the Blythe reference was not physically possible, for reasons described below, hence select deviations from the reported procedure were adopted. Being cognizant that significant deviation from the reported procedure could lead to compositional differences, I designed all deviations to be as minimally impactful to product reproduction as possible. For example, Example A-8 states:

"500 parts of textile spirits are then added, followed by 91.3 parts of concentrated nitric acid and 100 parts water. During the nitric acid addition the reaction temperature is maintained at 130 °-138 °C by external cooling."

While the text correctly infers that nitric acid addition is exothermic, it does call for maintaining a reaction temperature that greatly exceeds the boiling point of the bulk solvent, textile spirits, which has an average boiling point range of 66-85° C. This procedural contradiction could be rationalized if: a) the intention was to remove all lower-boiling species in the heating to 130° C or b) a typographical error exists such that either a different temperature and/or different solvent were intended. Upon careful consideration as to the least impactful compromise, I determined to co-add xylenes, a higher boiling solvent (b.p. 139° C). This would allow the reaction to reach the prescribed nitration temperature without incurring undue viscosity increase resulting from solvent removal. In my opinion, not reaching the prescribed temperature, and/or inadequately mixing reagents due to a viscosity increase would have egregiously perturbed the nitration reaction, compared to simply adding a relatively inert higher-boiling solvent such as xylenes.

Example A-8 subsequently states:

"During the nitric acid addition the reaction temperature is maintained at 130 °C-138 °C by external cooling. The reaction mixture is then stirred for two hours at room temperature and 61.5 parts of polyethylene polyamine described in Example A is added slowly."

If this procedure were to have been literally followed, all volatile species would have been removed from the reaction mixture prior to cooling to room temperature and the subsequent addition of the polyethylene polyamine. In the absence of any fluidizing

solvent(s), this material would have been too viscous to stir at room temperature. To mitigate this problem, and to facilitate any potential reaction with the polyamine, I included the xylenes to maintain fluidity at room temperature. Again, I deemed this deviation from the reported procedure to be less impactful than increasing reaction temperature during polyamine addition or inadequately mixing the reagents at room temperature.

Taking into account both aforementioned deviations from the procedure outlined in Example 8-A, an account of my replication is as follows:

A representative polybutene-substituted phenol\* (1443 parts) comprising a polybutene peak molecular weight of 1319 g/mol was mixed with 10 parts of aqueous hydrochloric acid and 33 parts paraformaldehyde at 90 °C for 20 hours under nitrogen with intermittent storage at room temperature. Thereafter 500 parts of hexanes/heptanes (which is equivalent to “textile spirits”), followed by 91.3 parts of concentrated nitric acid, 100 parts water and 500 parts of xylenes were added, after which time the stirring reaction mixture was heated and maintained at 138 °C for 3 hours before being cooled to room temperature. The fluid mixture, devoid of volatiles except xylenes, was treated with 61.5 parts of commercial grade tetraethylenepentamine and was heated to 160 °C for 7 hours before being stripped (first under nitrogen then followed by vacuum) to remove volatile materials. The residue was filtered to produce the finished product.

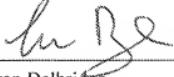
Samples of the finished product were given to Dr. Mosier for further testing.

We have been unable to unequivocally identify the chemical nature of the final product. Samples of both the “Intermediate” (before the addition of the tetraethylenepentamine) and the “Final Product” were submitted to our analytical group for analysis using Gel Permeation Chromatographic/Infrared Spectroscopic (GPC/IR) techniques. Spectra of the Intermediate revealed a weak, but definitive, absorption at 1538 cm<sup>-1</sup> which infers the presence of a nitro (NO<sub>2</sub>) group in the bulk material. In contrast, GPC/IR analysis of the final product appears devoid of this absorption at 1535 cm<sup>-1</sup> but was accompanied by the emergence of two new, broader absorptions around 1660 and 1593 cm<sup>-1</sup>. First inspection of these data might lead one to conclude that the nitrated Intermediate is subsequently “reduced” in the presence of the tetraethylenepentamine to afford an amino phenol in which the polyamine may or may not be attached. However,

\* This is a readily available Lubrizol stock material similar to that employed in Example A-8. The charge of this material was adjusted to provide the same number of equivalents (1.1) as that documented in Example A-8. All other charges were identical to those documented in Example A-8.

reduction of the nitro-phenol to an amino-phenol requires a reducing agent<sup>\*</sup> (metals; hydrazine *etc.*) of which no apparent source exists in the above procedure or Example A-8. In particular, amines/polyamines would not be anticipated to behave as a reducing agent under those reaction conditions. Additionally, the relatively broad absorption at 1593 cm<sup>-1</sup> in the Final Product may be obscuring the relatively weaker 1535 cm<sup>-1</sup> absorption observed in the Intermediate or, alternatively, the nitro absorption may have shifted due to the presence of polyamine. In other words it is difficult to infer from these data whether nitro reduction to an amine functional group has occurred to afford an aminophenol or whether any other reactions have occurred.

I further declare that all statements herein made of my own knowledge are true and all statements herein made on information and belief are believed to be true. I understand that willful false statements and the like are punishable by fine or imprisonment or both (18 U.S.C. 1001) and may jeopardize the validity of the application or any patent issuing thereon.



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Ewan Delbridge

12-15-09 (date)

<sup>\*</sup> Smith, M. B.; March, J.; *March's Advanced Organic Chemistry*, 5<sup>th</sup> Edition; Wiley Interscience, p. 1553.